

FORM PTO-1390 (REV. 5-93)	U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER 10191/1897
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		U.S. APPLICATION NO. (If known, see 37 CFR 1.5) <div style="font-size: 1.5em; font-weight: bold; text-align: center;">09/913482</div>
INTERNATIONAL APPLICATION NO. PCT/DE00/04149	INTERNATIONAL FILING DATE 23 November 2000 (23.11.00)	PRIORITY DATE CLAIMED: 15 December 1999 (15.12.99)
TITLE OF INVENTION ELECTROCHEMICAL SENSOR		
APPLICANT(S) FOR DO/EO/US Lothar DIEHL		
<p>Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information.</p> <ol style="list-style-type: none"> 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)) immediately rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1). 4. <input type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) <ol style="list-style-type: none"> a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US) <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)). 7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). b. <input type="checkbox"/> have been transmitted by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input checked="" type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)) (unsigned). 10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). <p>Items 11. to 16. below concern other document(s) or information included:</p> <ol style="list-style-type: none"> 11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 13. <input checked="" type="checkbox"/> A FIRST preliminary amendment. 14. <input checked="" type="checkbox"/> A substitute specification. 15. <input type="checkbox"/> A change of power of attorney and/or address letter. 16. <input checked="" type="checkbox"/> Other items or information: International Search Report (translated), and PCT/RO/101. 		

EXPRESS MAIL NO.: EL244507162US

- 17.
- ☒
- The following fees are submitted:

Basic National Fee (37 CFR 1.492(a)(1)-(5)):Search Report has been prepared by the EUROPEAN PATENT OFFICE or
JPO \$860.00

International preliminary examination fee paid to USPTO (37 CFR 1.482) \$690.00

No international preliminary examination fee paid to USPTO (37 CFR 1.482) but
international search fee paid to USPTO (37 CFR 1.445(a)(2)) \$710.00Neither international preliminary examination fee (37 CFR 1.482) nor international search
fee (37 CFR 1.445(a)(2)) paid to USPTO \$1,000.00International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims
satisfied provisions of PCT Article 33(2)-(4) \$100.00

CALCULATIONS | PTO USE ONLY

ENTER APPROPRIATE BASIC FEE AMOUNT =

\$ 860

Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30 months
from the earliest claimed priority date (37 CFR 1.492(e)).

\$

Claims	Number Filed	Number Extra	Rate		
Total Claims	23 - 20 =	3	X \$18.00	\$ 54	
Independent Claims	3 - 3 =	0	X \$80.00	\$ 0	
Multiple dependent claim(s) (if applicable)			+ \$270.00	\$	

TOTAL OF ABOVE CALCULATIONS =

\$ 914

Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must
also be filed. (Note 37 CFR 1.9, 1.27, 1.28).

\$

SUBTOTAL =

\$ 914

Processing fee of \$130.00 for furnishing the English translation later the ☐ 20 ☐ 30
months from the earliest claimed priority date (37 CFR 1.492(f)).

+

\$

TOTAL NATIONAL FEE =

\$ 914

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be
accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property

+

\$

TOTAL FEES ENCLOSED =

\$ 914

Amount to be:

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- a. ☐ A check in the amount of \$_____ to cover the above fees is enclosed.
- b. ☒ Please charge my Deposit Account No. 11-0600 in the amount of **\$914.00** to cover the above fees. A duplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 11-0600. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.SEND ALL CORRESPONDENCE TO:
Kenyon & Kenyon
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New York, New York 10004

SIGNATURE

Richard L. Mayer, Reg. No. 22,490
NAME

DATE

8/15/01

09/913482

[10191/1897]

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s) : Lothar DIEHL
Serial No. : To Be Assigned
Filed : Herewith
For : ELECTROCHEMICAL SENSOR
Art Unit : To Be Assigned
Examiner : To Be Assigned

Assistant Commissioner
for Patents
Washington, D.C. 20231
Box Patent Application

**PRELIMINARY AMENDMENT AND
37 C.F.R. § 1.125 SUBSTITUTE SPECIFICATION STATEMENT**

SIR:

Please amend the above-identified application before examination, as set forth below.

IN THE SPECIFICATION AND ABSTRACT:

In accordance with 37 C.F.R. § 1.121(b)(3), a Substitute Specification (including the Abstract, but without claims) accompanies this response. It is respectfully requested that the Substitute Specification (including Abstract) be entered to replace the Specification of record.

IN THE CLAIMS:

On the first page of the claims, first line, change "What is claimed is:" to:
--What Is Claimed Is--.

Please cancel original claims 1 to 21, without prejudice, in the underlying PCT
Application No. PCT/DE00/04149.

EL244507162us

Please add the following new claims:

22. (New) An electrochemical sensor for determining at least one of a gas component and a gas concentration in a gas mixture, comprising:
an ion-conducting solid electrolyte body;
at least one electrode situated on the ion-conducting solid electrolyte body; and
an electrode lead leading to the at least one electrode, wherein:
the electrode lead includes a material that possesses one of no ionic conductivity and an ionic conductivity that is significantly less than that of a material of the at least one electrode.
23. (New) The electrochemical sensor according to claim 22, wherein:
the at least one electrode and the electrode lead are each formed from a cermet material, and
a ceramic component of the at least one electrode is different than a ceramic component of the electrode lead.
24. (New) The electrochemical sensor according to claim 23, wherein:
the ceramic component of the electrode lead contains 5-10% by volume Al_2O_3 .
25. (New) The electrochemical sensor according to claim 23, wherein:
the ceramic component of the electrode contains 10-60% by volume ZrO_2 stabilized with Y_2O_3 .
26. (New) The electrochemical sensor according to claim 25, wherein:
the ceramic component of the electrode contains 20% by volume ZrO_2 stabilized with Y_2O_3 .
27. (New) The electrochemical sensor according to claim 25, wherein:
the at least one electrode includes an increased porosity as a result of adding a pore-forming material.

28. (New) The electrochemical sensor according to claim 23, wherein:
at least one of a metallic component of the at least one electrode and a metallic component of the electrode lead includes Pt.
29. (New) The electrochemical sensor according to claim 22, further comprising:
a wedge-shaped junction region including an overlap zone and being formed between the electrode lead and the at least one electrode.
30. (New) The electrochemical sensor according to claim 22, further comprising:
a heater; and
a layer plane in which the heater embedded in the ion-conducting solid electrolyte body is located, wherein:
at least one of the electrode lead and the at least one electrode is situated in the layer plane.
31. (New) The electrochemical sensor according to claim 30, wherein:
the heater is made of a material that is the same as the material of the electrode lead.
32. (New) The electrochemical sensor according to claim 22, wherein:
the at least one electrode includes at least one of an internal pump electrode and a reference electrode including corresponding electrode leads of a measuring cell.
33. (New) An electrochemical sensor for determining at least one of a gas component and a gas concentration in a gas mixture, comprising:
an ion-conducting solid electrolyte body;
at least one electrode situated on the ion-conducting solid electrolyte body; and
an electrode lead leading to the at least one electrode, wherein:
the electrode lead includes a material having a low resistance in comparison with a material of the at least one electrode.

34. (New) The electrochemical sensor according to claim 33, wherein:
the at least one electrode and the electrode lead are each formed from a cermet material, and
a ceramic component of the at least one electrode is different than a ceramic component of the electrode lead.
35. (New) The electrochemical sensor according to claim 34, wherein:
the ceramic component of the electrode lead contains 5-10% by volume Al_2O_3 .
36. (New) The electrochemical sensor according to claim 34, wherein:
the ceramic component of the electrode contains 10-60% by volume ZrO_2 stabilized with Y_2O_3 .
37. (New) The electrochemical sensor according to claim 36, wherein:
the ceramic component of the electrode contains 20% by volume ZrO_2 stabilized with Y_2O_3 .
38. (New) The electrochemical sensor according to claim 36, wherein:
the at least one electrode includes an increased porosity as a result of adding a pore-forming material.
39. (New) The electrochemical sensor according to claim 34, wherein:
at least one of a metallic component of the at least one electrode and a metallic component of the electrode lead includes Pt.
40. (New) The electrochemical sensor according to claim 33, further comprising:
a wedge-shaped junction region including an overlap zone and being formed between the electrode lead and the at least one electrode.
41. (New) The electrochemical sensor according to claim 33, further comprising:
a heater; and

a layer plane in which the heater embedded in the ion-conducting solid electrolyte body is located, wherein:

at least one of the electrode lead and the at least one electrode is situated in the layer plane.

42. (New) The electrochemical sensor according to claim 41, wherein:
the heater is made of a material that is the same as the material of the electrode lead.
43. (New) The electrochemical sensor according to claim 33, wherein:
the at least one electrode includes at least one of an internal pump electrode and a reference electrode including corresponding electrode leads of a measuring cell.
44. (New) An electrochemical sensor for determining at least one of a gas component and a gas concentration in a gas mixture, comprising:
an ion-conducting solid electrolyte body;
at least one electrode situated on the ion-conducting solid electrolyte body; and
an electrode lead leading to the at least one electrode, wherein:
the electrode lead includes a material having a low resistance in comparison with a material of the at least one electrode, and
the material possesses one of no ionic conductivity and an ionic conductivity that is significantly less in comparison with the material of the at least one electrode.

Remarks

This Preliminary Amendment cancels original claims 1 to 21 without prejudice, in the underlying PCT Application No. PCT/DE00/04149. The Preliminary Amendment also adds new claims 22-44. The new claims conform the claims to U.S. Patent and Trademark Office rules and do not add new matter to the application.

In accordance with 37 C.F.R. § 1.121(b)(3), the Substitute Specification (including the Abstract, but without the claims) contains no new matter. The amendments reflected in the Substitute Specification (including Abstract) are to conform the Specification and Abstract to U.S. Patent and Trademark Office rules or to correct informalities. As required by 37 C.F.R. § 1.121(b)(3)(iii) and § 1.125(b)(2), a Marked Up Version Of The Substitute Specification comparing the Specification of record and the Substitute Specification also accompanies this Preliminary Amendment. Approval and entry of the Substitute Specification (including Abstract) are respectfully requested.

The underlying PCT Application No. PCT/DE00/04149 includes an International Search Report, dated April 18, 2001, a copy of which is submitted herewith.

Applicant asserts that the subject matter of the present application is new, non-obvious, and useful. Prompt consideration and allowance of the application are respectfully requested.

Respectfully Submitted,
KENYON & KENYON

Dated: 8/15/01

By: Richard L. Mayer
Richard L. Mayer
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3/PATS

ELECTROCHEMICAL SENSOR

Field Of The Invention

The present invention is based on an electrochemical sensor.

Background Information

The sensors of this species must be heated in the active range to temperatures of more than ca. 350°C to achieve the necessary ionic conductivity of the solid electrolyte body. To increase the measuring accuracy of the sensor, it is known to control and, if necessary, to adjust the operating temperature of the measuring cell, i.e., of the solid electrolyte body in the measuring region. To this end, it is known to assign a heating device to the sensor, the heating device being capable of being switched on and off as a function of an operating temperature measured at the measuring cell.

To determine the operating temperature of the measuring cell, it is known to apply an a.c. voltage to the sensor and to use a measuring device to determine a total alternating-current resistance made up of the conjugate impedances of the solid electrolyte body and of the corresponding electrodes and electrode leads. The temperature-dependent internal resistance of the solid electrolyte body in the measuring region and, as such, its temperature in the measuring region can be deduced from the total resistance.

In the known method, it is disadvantageous that the measuring device, which determines the temperature-dependent resistance of the solid electrolyte body, uses a constant resistance of the electrodes and the electrode leads as a baseline. However, the resistance of the electrode leads and the electrodes is subject to a relatively high degree of scatter due to manufacture.

The measuring device adds this not insignificant scatter error to a temperature-dependent change in the resistance of the solid electrolyte body in the measuring region and provides a corresponding faulty control signal for the heating device of the sensor. As a result, the sensor is adjusted to an incorrect operating temperature.

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It is further disadvantageous that, in the lead region, the solid electrolyte body forms an additional internal resistance that is connected in parallel to the internal resistance of the solid electrolyte body in the region of the electrodes (measuring region) and also makes a not insignificant contribution to the total resistance. If, in addition, the temperature in the lead region is higher than in the measuring region, the internal resistance of the solid electrolyte body in the lead region is reduced, and it makes a contribution to the total resistance that is dependent on the temperature of the solid electrolyte body in the lead region. As a result, the sensor is likewise adjusted to an incorrect operating temperature.

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To avoid the effect of the internal resistance in the lead region, it is known from German Published Patent Application No. 198 37 607 to provide the lead of an electrode opposite the lead region of the solid electrolyte body with an electrically insulating layer. This design has the disadvantage that the use of at least one insulating layer additionally requires at least one printing step and is, therefore, expensive from a standpoint of production engineering.

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Summary Of The Invention

In comparison with the related art, the electrochemical sensor according to the present invention has the advantage of an improved regulation of the operating temperature, thereby enabling the sensor to function more precisely and more uniformly.

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The present invention achieves that the internal resistance between the electrode leads situated on a solid electrolyte body is significantly higher than that between the electrodes in question. Thus, the contribution to the total resistance made by the internal resistance in the lead region of the solid electrolyte body, which is connected in parallel to the internal resistance in the measuring region of the solid electrolyte body, is significantly reduced. Thus, the influence of the internal resistance in the lead region on the temperature regulation

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is negligible. An additional advantage from a standpoint of production engineering is that by dispensing with an electrically insulating layer, a printing step is no longer necessary.

According to the present invention, the resistance of at least one electrode lead makes a small contribution to the total resistance. Furthermore, the electrode lead is made of a material having a smaller degree of processing scatter with respect to its resistance. Thus, the effect of the resistance of the electrode lead on the total resistance is smaller.

The present invention additionally improves the regulation of the operating temperature of the sensor.

Designing the internal pump electrode lead and/or the reference electrode lead using a material having a lesser ionic conductivity or no ionic conductivity in comparison with the electrode in question has the additional advantage that the resistive coupling of the particular electrode leads that can lead to a loading effect of the pump voltage on the measuring voltage of the sensor cell is prevented. As a result, the $\lambda=1$ -ripple is decreased or even prevented, thereby further improving the control dynamic response of the sensor.

An additional advantage results from designing the external pump electrode lead and/or the internal pump electrode lead using a material having a low resistance in comparison with the material of the electrode in question. As a result, the drop in the pump voltage in the external pump electrode lead and/or internal pump electrode is reduced, thereby improving pump function.

A particular embodiment of the present invention provides that the reference electrode lead is situated in the layer plane of the heater, thereby eliminating at least one printing step. In a further embodiment of the present invention, the heater and reference electrode lead are produced from the same material, thereby resulting in an additional advantage from a standpoint of production engineering.

Brief Description Of The Drawings

Figure 1 shows an exploded view of a first exemplary embodiment of a sensor.

Figure 2 shows an exploded view of an additional exemplary embodiment of a sensor.

5 Figure 3 shows a top view of an electrode including an electrode lead of a sensor.

Figure 4 shows a top view of an electrode including an electrode lead as well as a heater.

Detailed Description

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Figure 1 shows an electrochemical sensor for analyzing gases, in the form of a planar sensor element 10. Sensor element 10 including a measuring region 61 and a lead region 62 has electrical connection contacts 60, a first solid electrolyte foil 11 designated as a heating foil, an insulating layer 12, a heater 13, an additional insulating layer 14, a second solid electrolyte foil 20 designated as a reference gas duct foil, as well as a reference electrode 21 having a reference electrode lead 22. Formed in reference gas duct foil 20 is a reference gas duct 29, which is connected via an opening in the lead region to air as a reference gaseous atmosphere. Above reference electrode 21 and reference electrode lead 22, the sensor element further has a third solid electrolyte foil 23 designated as a measuring foil, a measuring electrode 26 including measuring electrode lead 27, as well as a porous protective layer 28.

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Figure 2 shows an additional exemplary embodiment of an electrochemical sensor for analyzing gases. This sensor is a so-called broadband probe having two cells 37, 38. First cell 37 is a concentration cell that functions according to the Nernst principle. The operating mode of first cell 37 corresponds with the sensor described in Figure 1. Therefore, the same reference numerals are used for the same elements in Figure 2. Second cell 38 is an electrochemical pump cell that is laminated together with first cell 37 and that cooperates with the concentration cell in a method known per se, according to the functional principle of the broadband probe. Situated in the junction region between first cell 37 and second cell 38 is an intermediate layer 35 and a filler layer 34 for forming a space (not further represented) for accommodating diffusion barrier 30. Second cell 38 has an internal pump electrode 31, including an internal pump electrode lead 32, a fourth solid electrolyte foil 33 designated as a

pump foil, an external pump electrode 40, including an external pump electrode lead 41, and a porous protective layer 42. Measuring electrode lead 27 and internal pump electrode lead 32 run together in lead region 62 of sensor element 10.

5 Figure 3 shows a large surface of a solid electrolyte foil 49 having an electrode 50 and an electrode lead 51, which can, for example, form measuring electrode 26, including measuring electrode lead 27, or reference electrode 21, including reference electrode lead 22, of the sensor shown in Figure 1. The electrode 50 shown in Figure 3, including electrode lead 51, can, for example, also represent external pump electrode 40, including external pump
10 electrode lead 41, internal pump electrode 31, including internal pump electrode lead 32, measuring electrode 26, including measuring electrode lead 27, or reference electrode 21, including reference electrode lead 22, of the sensor shown in Figure 2.

Electrode lead 51 is made of an electrically conductive material, preferably platinum, and has a ceramic component for mechanical stabilization of 7% by volume Al_2O_3 , for example. Electrode 50 is made of a catalytic material, preferably platinum, and a ceramic material, preferably 20% by volume ZrO_2 stabilized with Y_2O_3 . In an additional embodiment, electrode 50 further has a porosity produced by a pore-forming material. The junction
15 between electrode 50 and electrode lead 51 is produced by a wedge-shaped junction region 52 having an overlap zone. Electrode 50 and electrode lead 51 are produced according to a method known per se, e.g. by screen printing.

The described design can be used in any combination for every electrode shown in Figures 1 and 2 and for the respective electrode leads. It is conceivable to also use the described design
20 of electrode 50 including electrode lead 51 for other electrochemical sensors of this type.

In the exemplary embodiment for the broadband probe (Figure 2), internal pump electrode lead 32 and/or reference electrode lead 22 are produced using Al_2O_3 as the ceramic component to reduce the $\lambda=1$ -ripple. In comparison with the ZrO_2 stabilized with Y_2O_3 ,
25 which is suitable as the ceramic material for electrode 21, 31, the Al_2O_3 possesses no ionic conductivity. As a result, there is no ionic conduction between electrode leads 22, 32, thereby increasing the internal resistance in this region.

A further exemplary embodiment of a broadband probe (Figure 2) is that to reduce the drop in pump voltage in the lead region, external pump electrode lead 41 features a material having a low resistance in comparison with the material of external pump electrode 40. This is achieved in that the proportion of electrically conductive material, e.g. platinum, is higher in the cermet material of external pump electrode lead 41 than in external pump electrode 40.

Figure 4 represents an additional specific embodiment in which electrode 50 and electrode lead 51, including a junction region 52, are situated in a layer plane in which a heater 55 embedded in the solid electrolyte body is located. For this purpose, heater 55, electrode 50, and electrode lead 55 are pressed onto first insulation layer 12, for example. In a preferred embodiment, heater 55 is produced from the same material as electrode lead 51.

NY01 400783 v 1

Abstract Of The Disclosure

An electrochemical sensor for determining a gas concentration of a measuring gas using a sensor element that has at least one electrode situated on an ion-conducting solid electrolyte body, an electrode lead leading to the electrode. The electrode lead is made of a material possessing no ionic conductivity or an ionic conductivity that is significantly less in comparison with the material of the electrode, and/or having a low resistance.

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3 / PRTS

ELECTROCHEMICAL SENSOR

Field Of The Invention

5 The present invention is based on an electrochemical sensor. [according to the definition of the species in Patent Claim 1]

Background Information

10 The sensors of this species must be heated in the active range to temperatures of more than ca. 350°C to achieve the necessary ionic conductivity of the solid electrolyte body. To increase the measuring accuracy of the sensor, it is known to control and, if necessary, to adjust the operating temperature of the measuring cell, i.e., of the solid electrolyte body in the measuring region. To this end, it is known to assign a heating device to the sensor, the heating device being capable of being switched on and off as a function of an operating
15 temperature measured at the measuring cell.

20 To determine the operating temperature of the measuring cell, it is known to apply an a.c. voltage to the sensor and to use a measuring device to determine a total alternating-current resistance made up of the conjugate impedances of the solid electrolyte body and of the corresponding electrodes and electrode leads. The temperature-dependent internal resistance of the solid electrolyte body in the measuring region and, as such, its temperature in the measuring region can be deduced from the total resistance.

25 In the known method, it is disadvantageous that the measuring device, which determines the temperature-dependent resistance of the solid electrolyte body, uses a constant resistance of the electrodes and the electrode leads as a baseline. However, the resistance of the electrode

leads and the electrodes is subject to a relatively high degree of scatter due to manufacture. The measuring device adds this not insignificant scatter error to a temperature-dependent change in the resistance of the solid electrolyte body in the measuring region and provides a corresponding faulty control signal for the heating device of the sensor. As a result, the sensor is adjusted to an incorrect operating temperature.

It is further disadvantageous that, in the lead region, the solid electrolyte body forms an additional internal resistance that is connected in parallel to the internal resistance of the solid electrolyte body in the region of the electrodes (measuring region) and also makes a not insignificant contribution to the total resistance. If, in addition, the temperature in the lead region is higher than in the measuring region, the internal resistance of the solid electrolyte body in the lead region is reduced, and it makes a contribution to the total resistance that is dependent on the temperature of the solid electrolyte body in the lead region. As a result, the sensor is likewise adjusted to an incorrect operating temperature.

To avoid the effect of the internal resistance in the lead region, it is known from [DE] German Published Patent Application No. 198 37 607 [A1] to provide the lead of an electrode opposite the lead region of the solid electrolyte body with an electrically insulating layer. This design has the disadvantage that the use of at least one insulating layer additionally requires at least one printing step and is, therefore, expensive from a standpoint of production engineering.

Summary [of the] Of The Invention

In comparison with the related art, the electrochemical sensor according to the present invention [and having the characterizing features of the independent claims] has the advantage of an improved regulation of the operating temperature, thereby enabling the sensor to function more precisely and more uniformly.

The present invention [described by the characterizing features of Claim 1] achieves that the internal resistance between the electrode leads situated on a solid electrolyte body is significantly higher than that between the electrodes in question. Thus, the contribution to the

total resistance made by the internal resistance in the lead region of the solid electrolyte body, which is connected in parallel to the internal resistance in the measuring region of the solid electrolyte body, is significantly reduced. Thus, the influence of the internal resistance in the lead region on the temperature regulation is negligible. An additional advantage from a standpoint of production engineering is that by dispensing with an electrically insulating layer, a printing step is no longer necessary.

According to the present invention [described by the characterizing features of independent Claim 11], the resistance of at least one electrode lead makes a small contribution to the total resistance. Furthermore, the electrode lead is made of a material having a smaller degree of processing scatter with respect to its resistance. Thus, the effect of the resistance of the electrode lead on the total resistance is smaller.

The present invention [according to independent Claim 21, which represents a combination of the distinguishing features of the first and the second independent claims,] additionally improves the regulation of the operating temperature of the sensor.

[The measures set forth in the dependent claims make possible advantageous developments of the sensor recited in the independent claims.]

Designing the internal pump electrode lead and/or the reference electrode lead using a material having a lesser ionic conductivity or no ionic conductivity in comparison with the electrode in question has the additional advantage that the resistive coupling of the particular electrode leads that can lead to a loading effect of the pump voltage on the measuring voltage of the sensor cell is prevented. As a result, the $\lambda=1$ -ripple is decreased or even prevented, thereby further improving the control dynamic response of the sensor.

An additional advantage results from designing the external pump electrode lead and/or the internal pump electrode lead using a material having a low resistance in comparison with the material of the electrode in question. As a result, the drop in the pump voltage in the external pump electrode lead and/or internal pump electrode is reduced, thereby improving pump function.

A particular embodiment of the present invention provides that the reference electrode lead is situated in the layer plane of the heater, thereby eliminating at least one printing step. In a further embodiment of the present invention, the heater and reference electrode lead are produced from the same material, thereby resulting in an additional advantage from a standpoint of production engineering.

Brief Description Of The Drawings [of the Drawing

The present invention is explained in greater detail by the drawings and the subsequent description. The figures show:]

Figure 1 shows an exploded view of a first exemplary embodiment of a sensor[;].

Figure 2 shows an exploded view of an additional exemplary embodiment of a sensor[;].

Figure 3 shows a top view of an electrode including an electrode lead of a sensor[; and].

Figure 4 shows a top view of an electrode including an electrode lead as well as a heater.

Detailed Description [of the Exemplary Embodiments]

Figure 1 shows an electrochemical sensor for analyzing gases, in the form of a planar sensor element 10. Sensor element 10 including a measuring region 61 and a lead region 62 has electrical connection contacts 60, a first solid electrolyte foil 11 designated as a heating foil, an insulating layer 12, a heater 13, an additional insulating layer 14, a second solid electrolyte foil 20 designated as a reference gas duct foil, as well as a reference electrode 21 having a reference electrode lead 22. Formed in reference gas duct foil 20 is a reference gas duct 29, which is connected via an opening in the lead region to air as a reference gaseous atmosphere. Above reference electrode 21 and reference electrode lead 22, the sensor element further has a third solid electrolyte foil 23 designated as a measuring foil, a measuring electrode 26 including measuring electrode lead 27, as well as a porous protective layer 28.

Figure 2 shows an additional exemplary embodiment of an electrochemical sensor for

analyzing gases. This sensor is a so-called broadband probe having two cells 37, 38. First cell 37 is a concentration cell that functions according to the Nernst principle. The operating mode of first cell 37 corresponds with the sensor described in Figure 1. Therefore, the same reference numerals are used for the same elements in Figure 2. Second cell 38 is an electrochemical pump cell that is laminated together with first cell 37 and that cooperates with the concentration cell in a method known per se, according to the functional principle of the broadband probe. Situated in the junction region between first cell 37 and second cell 38 is an intermediate layer 35 and a filler layer 34 for forming a space (not further represented) for accommodating diffusion barrier 30. Second cell 38 has an internal pump electrode 31, including an internal pump electrode lead 32, a fourth solid electrolyte foil 33 designated as a pump foil, an external pump electrode 40, including an external pump electrode lead 41, and a porous protective layer 42. Measuring electrode lead 27 and internal pump electrode lead 32 run together in lead region 62 of sensor element 10.

Figure 3 shows a large surface of a solid electrolyte foil 49 having an electrode 50 and an electrode lead 51, which can, for example, form measuring electrode 26, including measuring electrode lead 27, or reference electrode 21, including reference electrode lead 22, of the sensor shown in Figure 1. The electrode 50 shown in Figure 3, including electrode lead 51, can, for example, also represent external pump electrode 40, including external pump electrode lead 41, internal pump electrode 31, including internal pump electrode lead 32, measuring electrode 26, including measuring electrode lead 27, or reference electrode 21, including reference electrode lead 22, of the sensor shown in Figure 2.

Electrode lead 51 is made of an electrically conductive material, preferably platinum, and has a ceramic component for mechanical stabilization of 7% by volume Al_2O_3 , for example. Electrode 50 is made of a catalytic material, preferably platinum, and a ceramic material, preferably 20% by volume ZrO_2 stabilized with Y_2O_3 . In an additional embodiment, electrode 50 further has a porosity produced by a pore-forming material. The junction between electrode 50 and electrode lead 51 is produced by a wedge-shaped junction region 52 having an overlap zone. Electrode 50 and electrode lead 51 are produced according to a method known per se, e.g. by screen printing.

The described design can be used in any combination for every electrode shown in Figures 1 and 2 and for the respective electrode leads. It is conceivable to also use the described design of electrode 50 including electrode lead 51 for other electrochemical sensors of this type.

5 In the exemplary embodiment for the broadband probe (Figure 2), internal pump electrode lead 32 and/or reference electrode lead 22 are produced using Al_2O_3 as the ceramic component to reduce the $\lambda=1$ -ripple. In comparison with the ZrO_2 stabilized with Y_2O_3 , which is suitable as the ceramic material for electrode 21, 31, the Al_2O_3 possesses no ionic conductivity. As a result, there is no ionic conduction between electrode leads 22, 32, thereby
10 increasing the internal resistance in this region.

A further exemplary embodiment of a broadband probe (Figure 2) is that to reduce the drop in pump voltage in the lead region, external pump electrode lead 41 features a material having a low resistance in comparison with the material of external pump electrode 40. This is achieved in that the proportion of electrically conductive material, e.g. platinum, is higher in the cermet material of external pump electrode lead 41 than in external pump electrode 40.

Figure 4 represents an additional specific embodiment in which electrode 50 and electrode lead 51, including a junction region 52, are situated in a layer plane in which a heater 55 embedded in the solid electrolyte body is located. For this purpose, heater 55, electrode 50, and electrode lead 55 are pressed onto first insulation layer 12, for example. In a preferred embodiment, heater 55 is produced from the same material as electrode lead 51.

Abstract Of The Disclosure

An [The present invention relates to an] electrochemical sensor for determining a gas concentration of a measuring gas using a sensor element that has at least one electrode [(50)]
5 situated on an ion-conducting solid electrolyte body, an electrode lead [(51)] leading to the electrode. The electrode lead [(51)] is made of a material possessing no ionic conductivity or an ionic conductivity that is significantly less in comparison with the material of the electrode[(50)], and/or having a low resistance.

10 [(Figure 3)]

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3/PATS

[10191/1897]

ELECTROCHEMICAL SENSOR

Background Information

The present invention is based on an electrochemical sensor according to the definition of the species in Patent Claim 1.

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The sensors of this species must be heated in the active range to temperatures of more than ca. 350°C to achieve the necessary ionic conductivity of the solid electrolyte body. To increase the measuring accuracy of the sensor, it is known to control and, if necessary, to adjust the operating temperature of the measuring cell, i.e., of the solid electrolyte body in the measuring region. To this end, it is known to assign a heating device to the sensor, the heating device being capable of being switched on and off as a function of an operating temperature measured at the measuring cell.

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To determine the operating temperature of the measuring cell, it is known to apply an a.c. voltage to the sensor and to use a measuring device to determine a total alternating-current resistance made up of the conjugate impedances of the solid electrolyte body and of the corresponding electrodes and electrode leads. The temperature-dependent internal resistance of the solid electrolyte body in the measuring region and, as such, its temperature in the measuring region can be deduced from the total resistance.

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In the known method, it is disadvantageous that the measuring device, which determines the temperature-dependent resistance of the solid electrolyte body, uses a constant resistance of the electrodes and the electrode leads as a baseline. However, the resistance of the electrode leads and the electrodes is subject to a relatively high degree of scatter due to manufacture.

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The measuring device adds this not insignificant scatter error to a temperature-dependent change in the resistance of the solid electrolyte body in the measuring region and provides a corresponding faulty control signal for the heating device of the sensor. As a result, the sensor

is adjusted to an incorrect operating temperature.

It is further disadvantageous that, in the lead region, the solid electrolyte body forms an additional internal resistance that is connected in parallel to the internal resistance of the solid electrolyte body in the region of the electrodes (measuring region) and also makes a not insignificant contribution to the total resistance. If, in addition, the temperature in the lead region is higher than in the measuring region, the internal resistance of the solid electrolyte body in the lead region is reduced, and it makes a contribution to the total resistance that is dependent on the temperature of the solid electrolyte body in the lead region. As a result, the sensor is likewise adjusted to an incorrect operating temperature.

To avoid the effect of the internal resistance in the lead region, it is known from DE 198 37 607 A1 to provide the lead of an electrode opposite the lead region of the solid electrolyte body with an electrically insulating layer. This design has the disadvantage that the use of at least one insulating layer additionally requires at least one printing step and is, therefore, expensive from a standpoint of production engineering.

Summary of the Invention

In comparison with the related art, the electrochemical sensor according to the present invention and having the characterizing features of the independent claims has the advantage of an improved regulation of the operating temperature, thereby enabling the sensor to function more precisely and more uniformly.

The invention described by the characterizing features of Claim 1 achieves that the internal resistance between the electrode leads situated on a solid electrolyte body is significantly higher than that between the electrodes in question. Thus, the contribution to the total resistance made by the internal resistance in the lead region of the solid electrolyte body, which is connected in parallel to the internal resistance in the measuring region of the solid electrolyte body, is significantly reduced. Thus, the influence of the internal resistance in the lead region on the temperature regulation is negligible. An additional advantage from a standpoint of production engineering is that by dispensing with an electrically insulating layer, a printing step is no longer necessary.

According to the invention described by the characterizing features of independent Claim 11, the resistance of at least one electrode lead makes a small contribution to the total resistance. Furthermore, the electrode lead is made of a material having a smaller degree of processing scatter with respect to its resistance. Thus, the effect of the resistance of the electrode lead on the total resistance is smaller.

The invention according to independent Claim 21, which represents a combination of the distinguishing features of the first and the second independent claims, additionally improves the regulation of the operating temperature of the sensor.

The measures set forth in the dependent claims make possible advantageous developments of the sensor recited in the independent claims.

Designing the internal pump electrode lead and/or the reference electrode lead using a material having a lesser ionic conductivity or no ionic conductivity in comparison with the electrode in question has the additional advantage that the resistive coupling of the particular electrode leads that can lead to a loading effect of the pump voltage on the measuring voltage of the sensor cell is prevented. As a result, the $\lambda=1$ -ripple is decreased or even prevented, thereby further improving the control dynamic response of the sensor.

An additional advantage results from designing the external pump electrode lead and/or the internal pump electrode lead using a material having a low resistance in comparison with the material of the electrode in question. As a result, the drop in the pump voltage in the external pump electrode lead and/or internal pump electrode is reduced, thereby improving pump function.

A particular embodiment of the present invention provides that the reference electrode lead is situated in the layer plane of the heater, thereby eliminating at least one printing step. In a further embodiment of the present invention, the heater and reference electrode lead are produced from the same material, thereby resulting in an additional advantage from a standpoint of production engineering.

Brief Description of the Drawing

The present invention is explained in greater detail by the drawings and the subsequent description. The figures show:

- Figure 1 shows an exploded view of a first exemplary embodiment of a sensor;
Figure 2 shows an exploded view of an additional exemplary embodiment of a sensor;
Figure 3 shows a top view of an electrode including an electrode lead of a sensor; and
Figure 4 shows a top view of an electrode including an electrode lead as well as a heater.

Description of the Exemplary Embodiments

Figure 1 shows an electrochemical sensor for analyzing gases, in the form of a planar sensor element 10. Sensor element 10 including a measuring region 61 and a lead region 62 has electrical connection contacts 60, a first solid electrolyte foil 11 designated as a heating foil, an insulating layer 12, a heater 13, an additional insulating layer 14, a second solid electrolyte foil 20 designated as a reference gas duct foil, as well as a reference electrode 21 having a reference electrode lead 22. Formed in reference gas duct foil 20 is a reference gas duct 29, which is connected via an opening in the lead region to air as a reference gaseous atmosphere. Above reference electrode 21 and reference electrode lead 22, the sensor element further has a third solid electrolyte foil 23 designated as a measuring foil, a measuring electrode 26 including measuring electrode lead 27, as well as a porous protective layer 28.

Figure 2 shows an additional exemplary embodiment of an electrochemical sensor for analyzing gases. This sensor is a so-called broadband probe having two cells 37, 38. First cell 37 is a concentration cell that functions according to the Nernst principle. The operating mode of first cell 37 corresponds with the sensor described in Figure 1. Therefore, the same reference numerals are used for the same elements in Figure 2. Second cell 38 is an electrochemical pump cell that is laminated together with first cell 37 and that cooperates with the concentration cell in a method known per se, according to the functional principle of the broadband probe. Situated in the junction region between first cell 37 and second cell 38 is an intermediate layer 35 and a filler layer 34 for forming a space (not further represented) for accommodating diffusion barrier 30. Second cell 38 has an internal pump electrode 31, including an internal pump electrode lead 32, a fourth solid electrolyte foil 33 designated as a

pump foil, an external pump electrode 40, including an external pump electrode lead 41, and a porous protective layer 42. Measuring electrode lead 27 and internal pump electrode lead 32 run together in lead region 62 of sensor element 10.

Figure 3 shows a large surface of a solid electrolyte foil 49 having an electrode 50 and an electrode lead 51, which can, for example, form measuring electrode 26, including measuring electrode lead 27, or reference electrode 21, including reference electrode lead 22, of the sensor shown in Figure 1. The electrode 50 shown in Figure 3, including electrode lead 51, can, for example, also represent external pump electrode 40, including external pump electrode lead 41, internal pump electrode 31, including internal pump electrode lead 32, measuring electrode 26, including measuring electrode lead 27, or reference electrode 21, including reference electrode lead 22, of the sensor shown in Figure 2.

Electrode lead 51 is made of an electrically conductive material, preferably platinum, and has a ceramic component for mechanical stabilization of 7% by volume Al_2O_3 , for example. Electrode 50 is made of a catalytic material, preferably platinum, and a ceramic material, preferably 20% by volume ZrO_2 stabilized with Y_2O_3 . In an additional embodiment, electrode 50 further has a porosity produced by a pore-forming material. The junction between electrode 50 and electrode lead 51 is produced by a wedge-shaped junction region 52 having an overlap zone. Electrode 50 and electrode lead 51 are produced according to a method known per se, e.g. by screen printing.

The described design can be used in any combination for every electrode shown in Figures 1 and 2 and for the respective electrode leads. It is conceivable to also use the described design of electrode 50 including electrode lead 51 for other electrochemical sensors of this type.

In the exemplary embodiment for the broadband probe (Figure 2), internal pump electrode lead 32 and/or reference electrode lead 22 are produced using Al_2O_3 as the ceramic component to reduce the $\lambda=1$ -ripple. In comparison with the ZrO_2 stabilized with Y_2O_3 , which is suitable as the ceramic material for electrode 21, 31, the Al_2O_3 possesses no ionic conductivity. As a result, there is no ionic conduction between electrode leads 22, 32, thereby increasing the internal resistance in this region.

A further exemplary embodiment of a broadband probe (Figure 2) is that to reduce the drop in pump voltage in the lead region, external pump electrode lead 41 features a material having a low resistance in comparison with the material of external pump electrode 40. This is achieved in that the proportion of electrically conductive material, e.g. platinum, is higher in the cermet material of external pump electrode lead 41 than in external pump electrode 40.

Figure 4 represents an additional specific embodiment in which electrode 50 and electrode lead 51, including a junction region 52, are situated in a layer plane in which a heater 55 embedded in the solid electrolyte body is located. For this purpose, heater 55, electrode 50, and electrode lead 55 are pressed onto first insulation layer 12, for example. In a preferred embodiment, heater 55 is produced from the same material as electrode lead 51.

NY01 400700 v 1

What is claimed is:

1. An electrochemical sensor for determining gas components and/or gas concentrations in gas mixtures, having at least one electrode situated on an ion-conducting solid electrolyte body, an electrode lead leading to the electrode, wherein the electrode lead (51) contains a material that possesses no ionic conductivity or an ionic conductivity that is significantly less in comparison with the material of the electrode (50).
2. The electrochemical sensor as recited in Claim 1, wherein the electrode (50) and the electrode lead (51) are each formed from a cermet material; and the essential ceramic components of electrode (50) and electrode lead (51) are different.
3. The electrochemical sensor as recited in Claim 2, wherein the ceramic component of the electrode lead (51) contains 5-10% by volume Al_2O_3 .
4. The electrochemical sensor as recited in Claim 2, wherein the ceramic component of the electrode (50) contains 10-60% by volume, preferably 20% by volume, ZrO_2 stabilized with Y_2O_3 .
5. The electrochemical sensor as recited in Claim 4, wherein at least the electrode (50) has an increased porosity as a result of adding a pore-forming material.
6. The electrochemical sensor as recited in Claim 2, wherein the metallic component of the electrode (50) and/or of the electrode lead (51) contains Pt.
7. The electrochemical sensor as recited in Claim 1, wherein a wedge-shaped junction region (52) having an overlap zone is formed between electrode lead (51) and electrode (50).

8. The electrochemical sensor as recited in Claim 1,
wherein electrode lead (51) and/or electrode (50) is/are situated in a layer plane in
which a heater (55) embedded in the solid electrolyte body is located.
9. The electrochemical sensor as recited in Claim 8,
wherein the heater (55) is made of the same material as the electrode lead (51).
10. The electrochemical sensor as recited in Claim 1,
wherein the electrode (50) is an internal pump electrode (31) and/or a reference
electrode (21) having the corresponding electrode leads (32, 22) of a measuring cell.
11. An electrochemical sensor for determining gas components and/or gas concentrations
in gas mixtures, having at least one electrode situated on an ion-conducting solid
electrolyte body, an electrode lead leading to the electrode,
wherein the electrode lead (51) contains a material having a low resistance in
comparison with the material of the electrode (50).
12. The electrochemical sensor as recited in Claim 11,
wherein the electrode (50) and the electrode lead (51) are each formed from a cermet
material; and the essential ceramic components of electrode (50) and electrode lead
(51) are different.
13. The electrochemical sensor as recited in Claim 12,
wherein the ceramic component of the electrode lead (51) contains 5-10% by volume
 Al_2O_3 .
14. The electrochemical sensor as recited in Claim 12,
wherein the ceramic component of the electrode (50) contains 10-60% by volume,
preferably 20% by volume, ZrO_2 stabilized with Y_2O_3 .
15. The electrochemical sensor as recited in Claim 14,
wherein at least the electrode (50) has an increased porosity as a result of adding a
pore-forming material.

16. The electrochemical sensor as recited in Claim 12,
wherein the metallic component of the electrode (50) and/or of the electrode lead (51)
contains Pt.
17. The electrochemical sensor as recited in Claim 11,
wherein a wedge-shaped junction region (52) having an overlap zone is formed
between electrode lead (51) and electrode (50).
18. The electrochemical sensor as recited in Claim 11,
wherein electrode lead (51) and/or electrode (50) is/are situated in a layer plane in
which a heater (55) embedded in the solid electrolyte body is located.
19. The electrochemical sensor as recited in Claim 18,
wherein the heater (55) is made of the same material as the electrode lead (51).
20. The electrochemical sensor as recited in Claim 11,
wherein the electrode (50), including electrode lead (51), is an external pump
electrode (40) and/or an internal pump electrode (31) having the corresponding
electrode lead (41, 32).
21. An electrochemical sensor for determining gas components and/or gas concentrations
in gas mixtures, having at least one electrode situated on an ion-conducting solid
electrolyte body, an electrode lead leading to the electrode,
wherein the electrode lead (51) contains a material having a low resistance in
comparison with the material of the electrode (50), and also possessing no ionic
conductivity or an ionic conductivity that is significantly less in comparison with the
material of the electrode (50).

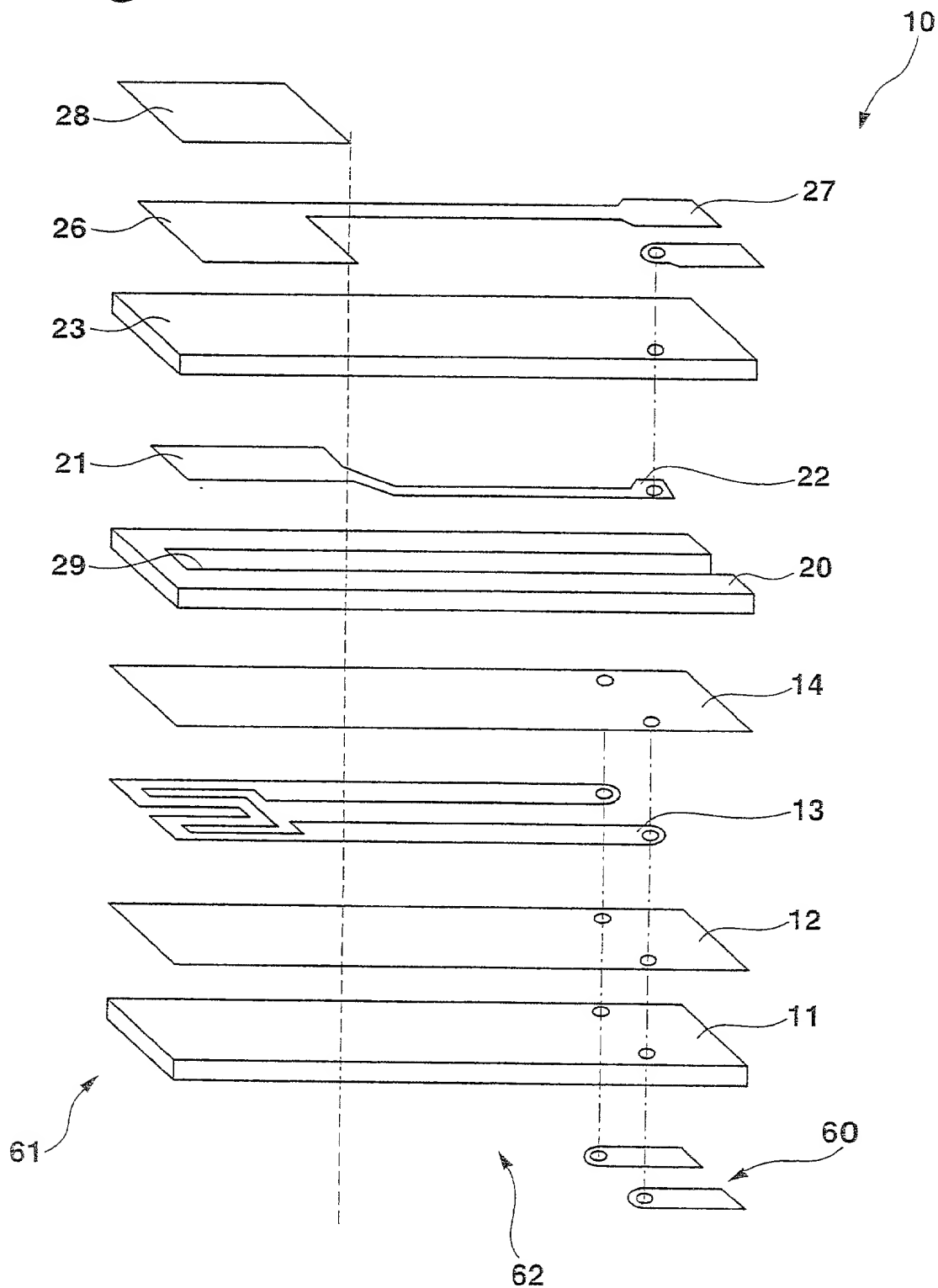
Fig. 1

Fig. 2

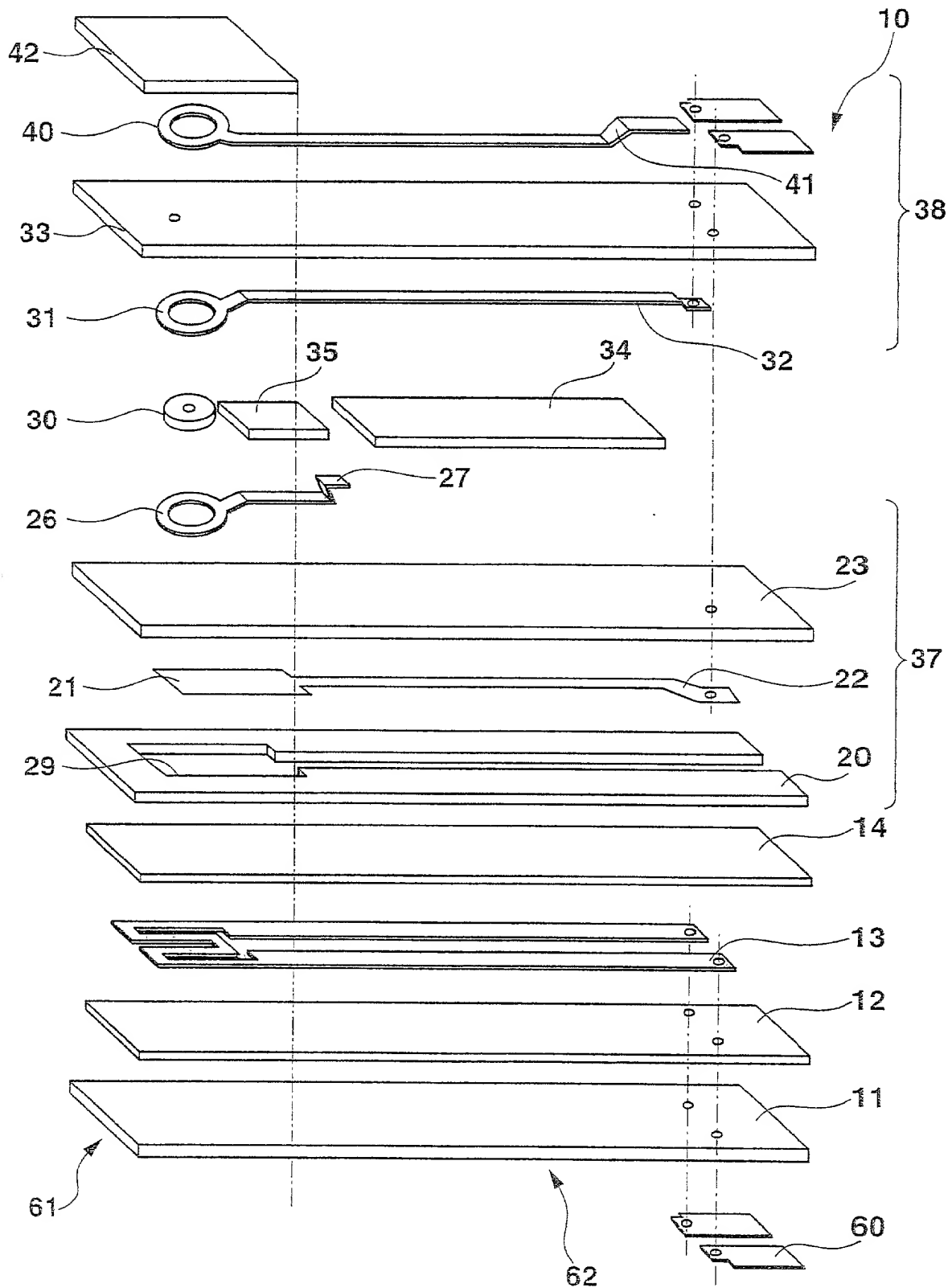
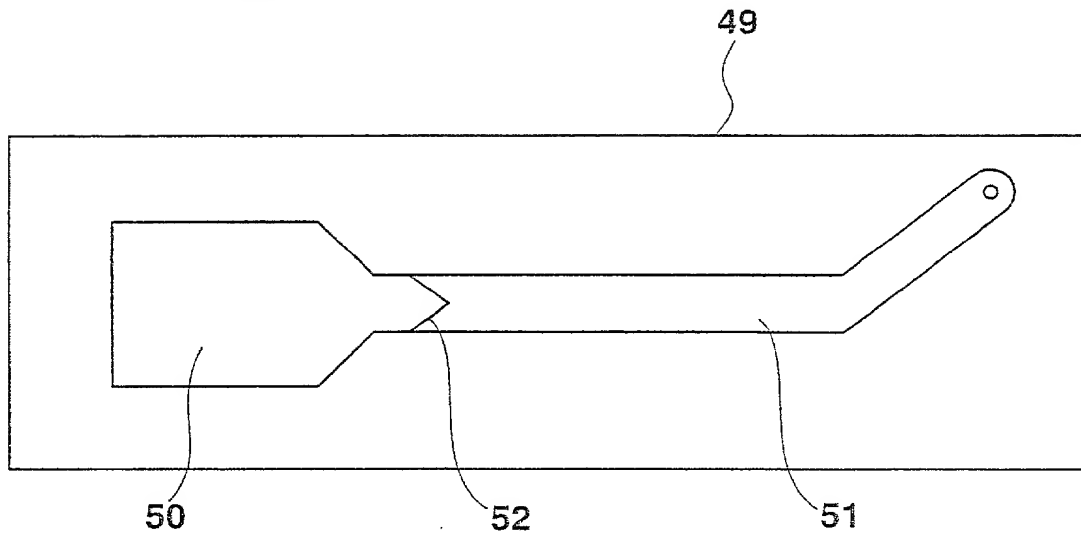
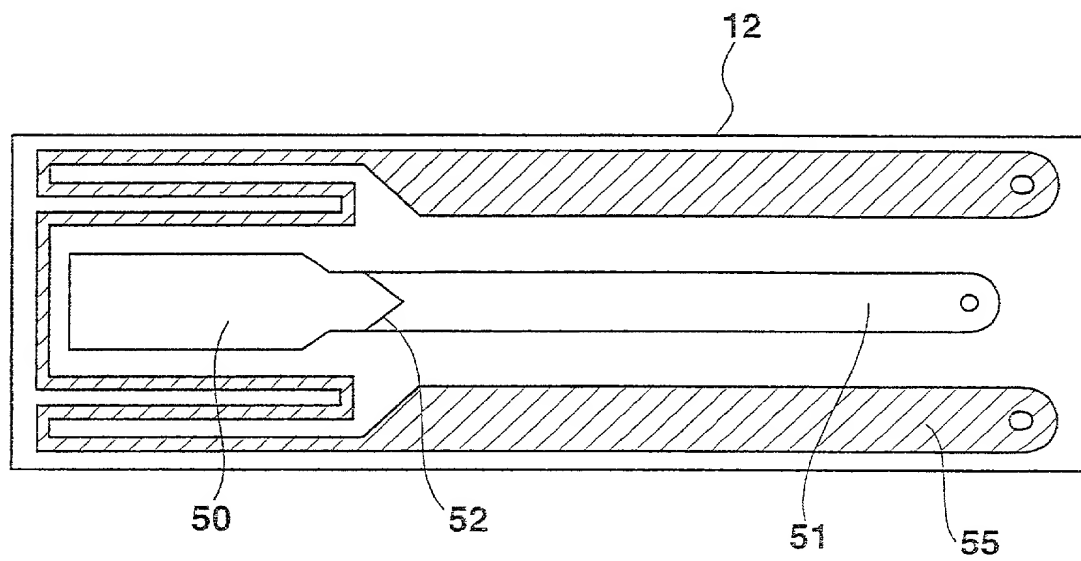


Fig. 3**Fig. 4**

**COMBINED DECLARATION AND
POWER OF ATTORNEY FOR PATENT APPLICATION**

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below adjacent to my name.

I believe I am the original, first and sole inventor of the subject matter which is claimed and for which a patent is sought on the invention entitled **ELECTROCHEMICAL SENSOR**, and the specification of which:

- ☐ is attached hereto;
- ☐ was filed as United States Application Serial No. _____ on _____, 19__ and was amended by the Preliminary Amendment filed on _____, 19__.
- ☒ was filed as PCT International Application Number PCT/DE00/04149 on the 23rd day of November, 2000.
- ☒ an English translation of which is filed herewith.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a). I hereby claim foreign priority benefits under Title 35, United States Code § 119 of any foreign application(s) for patent or inventor's certificate or of any PCT international applications(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

**PRIOR FOREIGN/PCT APPLICATION(S)
AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. § 119**

Country : Federal Republic of Germany

Application No. : 199 60 329.4

Date of Filing: December 15, 1999

Priority Claimed

Under 35 U.S.C. § 119 : ☒ Yes ☐ No

I hereby claim the benefit under Title 35, United States Code § 120 of any United States Application or PCT International Application designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code § 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations § 1.56(a) which occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application:

**PRIOR U.S. APPLICATIONS OR
PCT INTERNATIONAL APPLICATIONS
DESIGNATING THE U.S. FOR BENEFIT UNDER 35 U.S.C. § 120**

U.S. APPLICATIONS

Number :

Filing Date :

**PCT APPLICATIONS
DESIGNATING THE U.S.**

PCT Number :

PCT Filing Date :

I hereby appoint the following attorney(s) and/or agents to prosecute the above-identified application and transact all business in the Patent and Trademark Office connected therewith.

(List name(s) and registration number(s)):

(2)

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

1-00

Full name of inventor: Lothar DIEHL

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